CLAIMS

This listing of the claims will replace all prior versions, and listings of claims in the Application.

- 1. (currently amended) One-part self-etching, self-priming dental adhesive composition having a pH of at most 2, which comprises comprising:
 - (a) a polymerizable acidic phosphoric acid ester monomer of the following formula(A):

$$Y = \begin{pmatrix} 0 & 0 \\ 0 & 0 \end{pmatrix} \qquad (A)$$

wherein the moieties Y independent from each other represent a hydrogen atom or a moiety of the following formula (Y)

$$\left(z_{i}\right)$$
 O L (Y)

wherein

 Z_1 is COOR¹⁰, COSR²⁰, CON(R¹⁰)₂, CONR¹⁰R²⁰, or CONHR¹⁰, wherein R¹⁰ and R²⁰ independently represent

a hydrogen atom,

a C_{1-18} alkyl group optionally substituted by a C_{3-8} cycloalkyl group,

an optionally substituted C_{3-8} cycloalkyl group, an optionally substituted C_{4-18} aryl or heteroaryl group, an optionally substituted C_{5-18} alkylaryl or alkylheteroaryl group, or

an optionally substituted C₇₋₃₀ aralkyl group, whereby two R₁ residues may form together with the adjacent nitrogen atom to which they are bound a 5- to 7-membered heterocyclic ring which may contain further nitrogen atoms or an oxygen atoms,

and whereby the optionally substituted groups may be substituted by 1 to 5 C₁₋₅ alkyl groups; group(s);

L represents an (*a+b*)-valent organic residue, [[(]]whereby b is 1 when Y in formula (A) is within the round brackets[[is)]], L containing 2 to 45 carbon atoms and optionally heteroatoms, such as oxygen, nitrogen and sulfur atoms, the carbon atoms including *a + b* carbon atoms selected from primary and secondary aliphatic carbon atoms, secondary alicyclic carbon atoms, and aromatic carbon atoms, each of said *a+b* carbon atoms linking a phosphate or 2-(oxa-ethyl)acryl derivative group; a is an integer of from 1 to 10, preferably 1 to 5; b is an integer of from 1 to 10, preferably 1 to 5;

provided that at least one Y is not hydrogen; and

- (b) one or more polymerisable acidic monomers selected from the group consisting of
 - (b1) polymerisable acidic monomers of the following formula (B):

$$\begin{pmatrix}
R1 & N & L_1 & P & OH \\
R2 & C & OH
\end{pmatrix}_{d}$$
(B)

wherein

R₁ and R₂ independently represent

a hydrogen atom,

an optionally substituted C₁₋₁₈ alkyl group,

an optionally substituted C₃₋₁₈ cycloalkyl group,

an optionally substituted C₅₋₁₈ aryl or heteroaryl group,

an optionally substituted C₅₋₁₈ alkylaryl or alkylheteroaryl group,

an optionally substituted C₇₋₃₀ aralkyl group,

whereby the optionally substituted groups may be substituted by 1 to 5

C₁₋₅ alkyl groups; group(s);

L, represents a (c + d) valent organic residue containing 2 to 45 carbon atoms and optionally heteroatoms, such as oxygen, nitrogen and sulfur, the carbon atoms including c + d carbon atoms selected from primary and secondary aliphatic carbon atoms, secondary alicyclic carbon atoms, and aromatic carbon atoms, each of said c+d carbon atoms linking a phosphonate or optionally substituted acrylamido group;

and

c and d independently represent integers of from 1 to 10;

(b2) polymerisable acidic monomers of the following formula (C):

wherein

 Z_2 independently has the same meaning as defined for Z_1 ;

 L_2 represents an (e+f) valent organic residue containing 2 to 45 carbon atoms and optionally heteroatoms, such as oxygen, nitrogen and sulfur atoms, the carbon atoms including e+f carbon atoms selected from primary and secondary aliphatic carbon atoms, secondary alicyclic carbon atoms, and aromatic carbon atoms, each of said e+f carbon atoms linking a sulphonate or optionally substituted 2-(oxa-ethyl)acryl derivative group; and e and f independently represent an integer of from 1 to 10;

(b3) acidic monomers of the following formula (D):

wherein

R₃ and R₄ independently represent

a hydrogen atom,

an optionally substituted C₁₋₁₈ alkyl group,

an optionally substituted C₃₋₁₈ cycloalkyl group,

an optionally substituted C₅₋₁₈ aryl or heteroaryl group,

an optionally substituted C₅₋₁₈ alkylaryl or alkylheteroaryl group,

an optionally substituted C7-30 aralkyl group,

whereby the optionally substituted groups may be substituted by 1 to 5 C₁-

5 alkyl groups; group(s)

 L_3 represents a (g+h) valent valent organic residue containing 2 to 45 carbon atoms and optionally heteroatoms, such as oxygen, nitrogen and sulfur atoms, the carbon atoms including g+h carbon atoms selected from primary and secondary aliphatic carbon atoms, secondary alicyclic carbon atoms, and

aromatic carbon atoms, each of said g+h carbon atoms linking a sulphonate or optionally substituted acrylamido group; and g and h independently represent integers ef-from 1 to 10;

- (c) a polymerizable N-substituted alkylacrylic or acrylic acid amide monomer;
- (d) optionally an organic and/or inorganic acid;
- (e) an organic water soluble solvent and/or water; and
- (f) <u>a</u> polymerization initiator[[,]];
- (g) an inhibitor; and
- (h) <u>a</u> stabilizer.
- 2. (original) The one-part self-etching, self-priming dental adhesive composition of claim 1 wherein L₁, L₂, and L₃ independently represent an optionally substituted C₁₋₁₈ alkylene group which may contain from 1 to 9 oxygen atoms in the chain, an optionally substituted C₃₋₁₈ cycloalkylene group, an optionally substituted C₅₋₁₈ arylene or heteroarylene group, an optionally substituted C₅₋₁₈ alkylaryl or alkylheteroarylene group, an optionally substituted C₇₋₃₀ aralkylene group.
- 3. (currently amended) The one-part self-etching, self-priming dental adhesive composition of claim 1 or 2 which is hydrolysis stable for at least one week at a storage temperature of 50 °C, whereby after such storage the bond strength of an adhesive prepared from such an adhesive composition to enamel and/or dentin is at least 10 MPa, preferably 15 MPa.
- 4. (currently amended) The one-part self-etching, self-priming dental adhesive composition according to <u>claim 1</u>, <u>any one of the preceding claims</u>, wherein components (a) and (b) are contained in a ratio of from 1:100 to 100 : 1.
- 5. (currently amended) The one-part self-etching, self-priming dental adhesive composition according to <u>claim 1</u>, <u>any one of the preceding claims</u>, wherein said organic acid of component (d) is selected from the group <u>consisting</u> of mono- or polycarboxylic acids, <u>such as methacrylic acid</u>, acrylic acid, fumaric acid, maleic acid, citric acid, itaconic acid,

<u>and formic acid</u>, and wherein the inorganic acid of component (d) is selected from the group <u>consisting</u> of sulfonic acid, phosphoric acid, sulfuric acid and hydrofluoric acid.

- 6. (currently amended) The one-part self-etching, self-priming dental adhesive composition according to <u>claim 1</u>, <u>any one of the preceding claims</u>, wherein said organic water soluble solvent of component (e) is selected from the group <u>consisting</u> of alcohols, <u>and</u> ketones, <u>such as ethanol</u>, propanol, butanol, acetone, <u>and methyl ethyl ketone</u>.
- 7. (currently amended) The one-part self-etching, self-priming dental adhesive composition according to <u>claim 1</u>, <u>any one of the preceding claims</u>, wherein said acidic polymerizable monomer of component (a) is characterized by one of the following formulas:

wherein

Z or Z_1 is COOR¹⁰, COSR²⁰, CON(R¹⁰)₂, CONR¹⁰R²⁰, or CONHR¹⁰, wherein R¹⁰ and R²⁰ independently represent a hydrogen atom,

a C_{1^-18} alkyl group optionally substituted by a C_{3^-8} cycloalkyl group, an optionally substituted C_{3^-8} cycloalkyl group, an optionally substituted C_{4^-18} aryl or heteroaryl group, an optionally substituted C_{5^-18} alkylaryl or alkylheteroaryl group, or an optionally substituted C_{7^-30} aralkyl group, whereby two R_1 residues may form together with the adjacent nitrogen atom to which they are bound a 5- to 7-membered heterocyclic ring which may contain further nitrogen atoms or an oxygen atoms,

and whereby the optionally substituted groups may be substituted by 1 to 5 C₁₋₅ alkyl groups; group(s);

L represents an (a+b)-valent organic residue containing 2 to 45 carbon atoms and optionally heteroatoms, such as oxygen, nitrogen and sulfur atoms, the carbon atoms including a+b carbon atoms selected from primary and secondary aliphatic carbon atoms, secondary alicyclic carbon atoms, and aromatic carbon atoms, each of said a+b carbon atoms linking a phosphate or 2-(oxa-ethyl)acryl derivative group;

a is an integer of from 1 to 10, preferably 1 to 5;

b is an integer of from 1 to 10, preferably 1 to 5

wherein Z is as defined in claim 1 and n is an integer.

- 8. (currently amended) The one-part self-etching, self-priming dental adhesive composition according to <u>claim 1</u>, <u>any one of the preceding claims</u>, wherein said acidic polymerizable monomer of component (b) is a polymerisable acidic monomers of formula (C).
- 9. (currently amended) Hydrolysis stable <u>The</u> one-part self-etching, self-priming dental adhesive composition of claim 8, wherein said acidic polymerizable monomer is characterized by one of the following formulas:

10. (currently amended) The one-part self-etching, self-priming dental adhesive composition according to <u>claim 1</u>, <u>any one of the preceding claims</u>, wherein the polymerizable N-substituted alkylacrylic or acrylic acid amide monomer of component (c) is characterized by one of the following formulas:

wherein

R₅ and R₆ independently represent

a hydrogen atom or a substituted

a C₁ to C₁₈ alkyl group,

an optionally substituted C₃₋₁₈ cycloalkyl group,

an optionally substituted C₅₋₁₈ aryl or heteroaryl group,

an optionally substituted C₅₋₁₈ alkylaryl or alkylheteroaryl group,

an optionally substituted C₇₋₃₀ aralkyl group,

R₇ represents a

a divalent substituted or unsubstituted organic residue having from 1 to 45 carbon atoms, whereby said organic residue may contain from 1 to 14 oxygen and/or nitrogen atoms and is selected from a C_1 to C_{18} alkylene group wherein from 1 to C_{18} are C_{18} groups may be replaced by a C_{18} and C_{18} group wherein C_{18} is a

hydrogen atom or a C_1 to C_{18} alkyl group, a divalent substituted or unsubstituted C_3 to C_{18} cycloalkyl or cycloalkylene group, a divalent substituted or unsubstituted C_4 to C_{18} aryl or heteroaryl group, a divalent substituted or unsubstituted C_5 to C_{18} alkylaryl or alkylheteroaryl group, a divalent substituted or unsubstituted C_7 to C_{30} aralkyl group, and a divalent substituted or unsubstituted C_2 to C_{45} mono-, di- or polyether group having from 1 to 14 oxygen atoms,

R₈ represents

a saturated di- or multivalent substituted or unsubstituted C_2 to C_{18} hydrocarbon group, a saturated di- or multivalent substituted or unsubstituted cyclic C_3 to C_{18} hydrocarbon group, a di- or multivalent substituted or unsubstituted C_4 to C_{18} aryl or heteroaryl group, a di- or multivalent substituted or unsubstituted C_5 to C_{18} alkylaryl or alkylheteroaryl group, a di- or multivalent substituted or unsubstituted C_7 to C_{30} aralkyl group, or a di- or multivalent substituted or unsubstituted C_2 to C_{45} mono-, di-, or polyether residue having from 1 to 14 oxygen atoms, and n is an integer.

11. (currently amended) The one-part self-etching, self-priming dental adhesive composition according to <u>claim 1</u>, <u>any one of the preceding claims</u>, wherein said polymerizable monomer is a mono-, bis- or poly(meth) acrylamide characterized by one of the following formulas:

- 12. (currently amended) The one-part self-etching, self-priming dental adhesive composition according to <u>claim 1</u>, <u>any one of the preceding claims</u>, which contains said acidic polymerizable monomers of components (a) and (b) in an amount <u>of</u> from 5 to 90 wt-%.
- 13. (currently amended) The one-part self-etching, self-priming dental adhesive composition according to <u>claim 1</u>, <u>any one of the preceding claims</u>, wherein said polymerization initiator is a thermal initiator, a redox-initiator or a photo initiator.
- 14. (currently amended) The one-part self-etching, self-priming dental adhesive composition according to <u>claim 13</u>, any one of the preceding claims, wherein said photo initiator is champhor quinone.
- 15. (currently amended) The one-part self-etching, self-priming dental adhesive composition according to <u>claim 1</u>, <u>any one of the preceding claims</u>, wherein said filler is an inorganic filler and/or an organic filler; <u>preferably the filler is a nanofiller</u>.
- 16. (currently amended) The one-part self-etching, self-priming dental adhesive composition according to <u>claim 1</u>, <u>any-one-of the preceding claims</u>, wherein said stabilizer is a radical absorbing monomer, <u>such as hydroquinone</u>, hydroquinone monomethylether, 2,6-di-tert.-butyl-p-cresol.

17. (currently amended) The one-part self-etching, self-priming dental adhesive composition according to <u>claim 1</u>, any one of the preceding claims, wherein L represents

an (a+b)-valent saturated aliphatic C_2 to C_{18} group having at least 2 of said primary aliphatic carbon atoms, and optionally 1 or more of said secondary aliphatic carbon atom(s), whereby said (a+b)-valent group may be substituted by C_1 to C_5 alkyl group(s); or

a C_2 to C_{45} mono-, di-, or polyether which has from 1 to 14 oxygen atoms and is substituted by at least 2 C_1 to C_{10} aliphatic <u>groups group(s)</u> having said primary and/or secondary aliphatic carbon atoms; whereby said ether may optionally be substituted by C_1 to C_5 alkyl <u>groups</u>; group(s); or

wherein L represents:

a saturated C_3 to C_8 cyclic, C_7 to C_{15} bi- or polycyclic hydrocarbon group having from 0 to 4, preferably, 0 to 3, more preferably 0 or 1, of said secondary alicyclic carbon atoms; and/or

a C_4 to C_{18} aryl or heteroaryl group having from 0 to 5, preferably 0 to 3, more preferably 0 or 1, of said aromatic carbon atoms; whereby said saturated hydrocarbon or aryl or heteroaryl group is substituted by

from 0 to 5 C₁ to C₅ alkyl groups; group(s);

from 0 to 4, preferably 1 to 3, more preferably 1 or 2, saturated C_1 to C_{10} aliphatic group(s) having said primary and/or secondary aliphatic carbon atoms, and/or from 0 to 2 divalent residues according to one of the following formulas:

-[O-CH₂CH₂-]_f wherein f is an integer of from 1 to 10, preferably 1 to 5;

-[-O-CH $_2$ CH $_2$ CH $_2$ -] $_g$ - wherein g is an integer from offrom 1 to 10, preferably 1 to 5;

- $[O-R_{12}]_{h}$ - wherein R_{12} is -CH(CH₃)-CH₂- or -CH₂-CH(CH₃)- and h is an integer of from 1 to 10, preferably 1-to 5;

-[-O-R₁₄]_i-[O-R₁₅]_j- or -[O-R₁₅]_k-[O-R₁₄]_l- wherein R₁₄ is -CH₂CH₂-, R₁₅ is -CH(CH₃)-CH₂- or -CH₂-CH(CH₃)-, i, j, k, and I are integers whereby

 $2i + 3j \le 15$ and $2k + 3l \le 15$,

-[O-CH₂CH₂CH₂CH₂-]_r- wherein r is an integer of 1 or 2;

wherein said divalent residues have one of said primary aliphatic carbon atoms; and

whereby 2 groups selected from said saturated hydrocarbon, aryl, and heteroaryl groups may optionally be linked by a single bond, an alkylene group, or -O-.

- 18. (currently amended) The one-part self-etching, self-priming dental adhesive composition according to claim 1, any one of the preceding claims, wherein L represents an (a+b)-valent saturated C₃ to C₈ cyclic or C₇ to C₁₅ bi- or tricyclic hydrocarbon group having at least 2 of said secondary alicyclic carbon atoms; an (a+b)-valent saturated C₄ to C₁₈ aryl or heteroaryl group having from 2 to 6 of said aromatic carbon atoms; an (a+b)-valent C₆ to C₁₈ alkylaryl or alkyl heteroaryl group having at least one of said aromatic carbon atoms, at least one of said secondary aliphatic carbon atoms, and optionally one of said primary aliphatic carbon atoms at the terminal end of the alkyl moiety of said alkylaryl or alkylheteroaryl group; or an (a+b)-valent C₈ to C₃₀ aralkyl group having at least one of said primary aliphatic carbon atoms and at least one of said secondary aliphatic carbon atoms.
- 19. (currently amended) The one-part self-etching, self-priming dental adhesive composition according to <u>claim 1</u>, <u>any one of the preceding claims</u>, wherein L represents is a divalent residue according to one of the following formulas:
 - -[CH₂CH₂-O-]_m-CH₂CH₂- wherein m is an integer of from 1 to 14,
 - -[CH₂CH₂CH₂-O-]_p-CH₂CH₂ CH₂- wherein p is an integer of from 1 to 14,
 - $-[R_{12}-O]_{\sigma}-R_{13}$ wherein R_{12} and R_{13} may be $-CH(CH_3)-CH_2$ or
 - -CH₂-CH(CH₃)- and q is from 1 to 14,
 - $-[R_{14}-O]_{t}-[R_{15}-O]_{s}-R_{14}- \text{ or } -[R_{14}-O]_{t}-[R_{15}-O]_{u}-R_{15}- \text{ wherein } R_{14} \text{ is}$
 - $-CH_2CH_{2^-}$, R_{15} is $-CH(CH_3)-CH_{2^-}$ or $-CH_2-CH(CH_3)-$, r, s, t, and u are integers whereby $2r + 3s \le 43$ and $2t + 3u \le 42$,
 - -[CH₂CH₂CH₂CH₂-O-]_r-CH₂CH₂CH₂CH₂- wherein r is 1 or 2,

wherein R_{16} and R_{17} are H or -CH₃ and x and y may independently be integers of from 0 to 10, preferably 0 to 5.

- 20. (currently amended) The one-part self-etching, self-priming dental adhesive composition according to <u>claim 1</u>, <u>any one of the preceding claims</u>, wherein said (*a*+*b*) carbon atoms are primary aliphatic carbon atoms.
- 21. (currently amended) The one-part self-etching, self-priming dental adhesive composition according to <u>claim 1</u>, <u>any one of the preceding claims</u>, wherein the polymerizable acidic phosphoric acid ester monomer is of the following formula (A-1):

$$\begin{pmatrix}
z_1 & O \\
D & O \\
HO & OH
\end{pmatrix}_b$$
(A-1)

wherein

or

 Z_1 is $COOR^{10}$, $COSR^{20}$, $CON(R^{10})_2$, $CONR^{10}R^{20}$, or $CONHR^{10}$, wherein

R¹⁰ and R²⁰ independently represent

a hydrogen atom,

a $C_{1^{-1}8}$ alkyl group optionally substituted by a $C_{3^{-8}}$ cycloalkyl group,

an optionally substituted C_{3^-8} cycloalkyl group, an optionally substituted C_{4^-18} aryl or heteroaryl group, an optionally substituted C_{5^-18} alkylaryl or alkylheteroaryl group, or an optionally substituted C_{7^-30} aralkyl group, whereby two R_1 residues may form together with the adjacent nitrogen atom to which they are bound a 5- to 7-membered heterocyclic ring which may contain further nitrogen atoms or an oxygen atoms,

and whereby the optionally substituted groups may be substituted by 1 to 5 C_{1-5} alkyl groups; group(s);

L represents an (a+b)-valent organic residue containing 2 to 45 carbon atoms and optionally heteroatoms, such as oxygen, nitrogen and sulfur atoms, the carbon atoms including a+b carbon atoms selected from primary and secondary aliphatic carbon atoms, secondary alicyclic carbon atoms, and aromatic carbon atoms, each of said a+b carbon atoms linking a phosphate or 2-(oxa-ethyl)acryl derivative group;

a is an integer of from 1 to 10, preferably 1 to 5; b is an integer of from 1 to 10, preferably 1 to 5.

- 22. (currently amended) The one-part self-etching, self-priming dental adhesive composition according to <u>claim 1</u>, <u>any one of claims 1 to 20</u> wherein none of the moieties Y is a hydrogen atom.
- 23. (currently amended) A polymerizable acidic phosphoric acid ester monomer of the following formula (A)

$$Y = \begin{pmatrix} 0 & 0 \\ 0 & 0 - Y \end{pmatrix}_{b}$$
 (A)

wherein

the moieties Y independent from each other represent a moiety of the following formula (Y)

wherein

 Z_1 is COOR¹⁰, COSR²⁰, CON(R¹⁰)₂, CONR¹⁰R²⁰, or CONHR¹⁰, wherein R¹⁰ and R²⁰ independently represent

a hydrogen atom,

a $C_{1^{-}18}$ alkyl group optionally substituted by a $C_{3^{-}8}$ cycloalkyl group,

an optionally substituted C_{3^-8} cycloalkyl group, an optionally substituted C_{4^-18} aryl or heteroaryl group, an optionally substituted C_{5^-18} alkylaryl or alkylheteroaryl group, or

an optionally substituted C_{7^-30} aralkyl group, whereby two R_1 residues may form together with the adjacent nitrogen atom to which they are bound a 5- to 7-membered heterocyclic ring which may contain further nitrogen atoms or an oxygen atoms,

and whereby the optionally substituted groups may be substituted by 1 to 5 C_{1-5} alkyl groups; group(s);

L represents an (a+b)-valent organic residue, [[(]]whereby b is 1 when Y in formula (A) is within the round brackets[[is)]], L containing 2 to 45 carbon atoms and optionally heteroatoms, such as oxygen, nitrogen and sulfur atoms, the carbon atoms including a+b carbon atoms selected from primary and secondary aliphatic carbon atoms, secondary alicyclic carbon atoms, and aromatic carbon atoms, each of said a+b carbon atoms linking a phosphate or 2-(oxa-ethyl)acryl derivative group;

a is an integer of from 1 to 10, preferably 1 to 5;

b is an integer of from 1 to 10, preferably 1 to 5, more preferably 1.